

## **Fuel-Cycle and Nuclear Material Disposition Issues Associated with High-Temperature Gas Reactors**

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### **Abstract**

The objective of this paper is to facilitate a better understanding of the fuel-cycle and nuclear material disposition issues associated with high-temperature gas reactors (HTGRs). This paper reviews the nuclear fuel cycles supporting early and present day gas reactors, and identifies challenges for the advanced fuel cycles and waste management systems supporting the next generation of HTGRs, including the Very High Temperature Reactor, which is under development in the Generation IV Program.

The earliest gas-cooled reactors were the carbon dioxide (CO<sub>2</sub>)-cooled reactors. Historical experience is available from over 1,000 reactor-years of operation from 52 electricity-generating, CO<sub>2</sub>-cooled reactor plants that were placed in operation worldwide. Following the CO<sub>2</sub> reactor development, seven HTGR plants were built and operated.

The HTGR came about from the combination of helium coolant and graphite moderator. Helium was used instead of air or CO<sub>2</sub> as the coolant. The helium gas has a significant technical base due to the experience gained in the United States from the 40-MW<sub>e</sub> Peach Bottom and 330-MW<sub>e</sub> Fort St. Vrain reactors designed by General Atomics. Germany also built and operated the 15-MW<sub>e</sub> Arbeitsgemeinschaft Versuchsreaktor (AVR) and the 300-MW<sub>e</sub> Thorium High-Temperature Reactor (THTR) power plants. The AVR, THTR, Peach Bottom and Fort St. Vrain all used fuel containing thorium in various forms (i.e., carbides, oxides, thorium particles) and mixtures with highly enriched uranium. The operational experience gained from these early gas reactors can be applied to the next generation of nuclear power systems. HTGR systems are being developed in South Africa, China, Japan, the United States, and Russia.

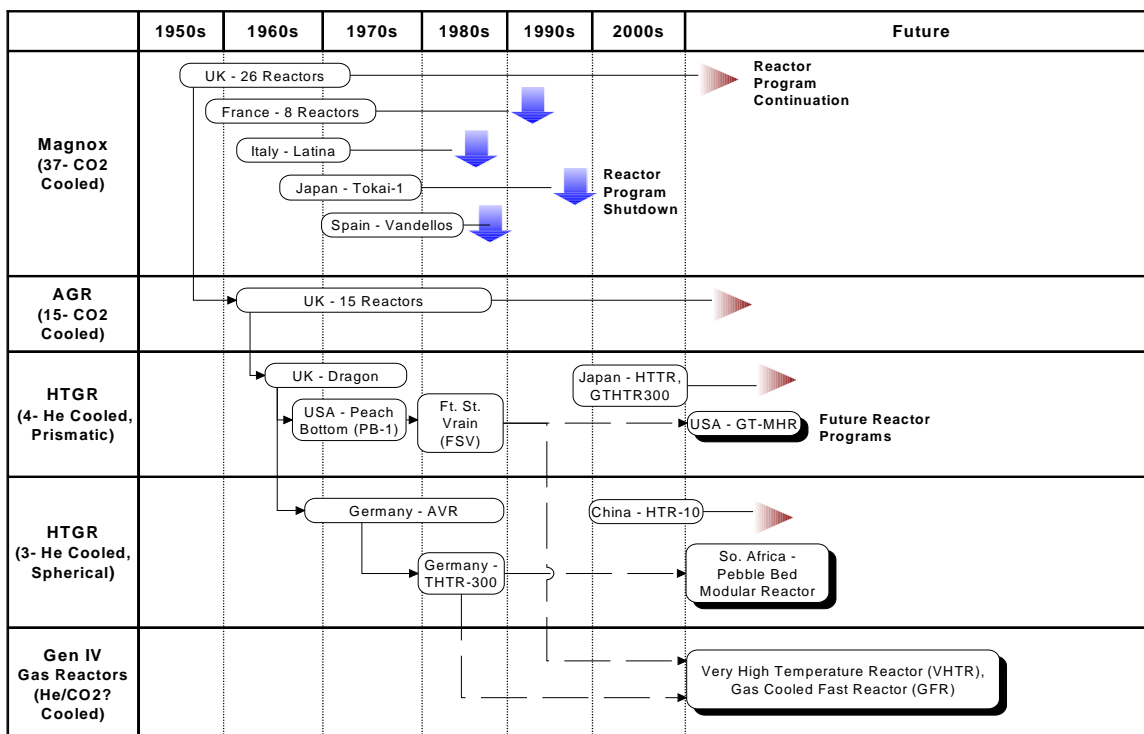
Elements of the HTGR system evaluated included fuel demands on uranium ore mining and milling, conversion, enrichment services, and fuel fabrication; fuel management in-core; spent fuel characteristics affecting fuel recycling and refabrication, fuel handling, interim storage, packaging, transportation, waste forms, waste treatment, decontamination and decommissioning issues; and low-level waste (LLW) and high-level waste (HLW) disposal.

## 1. Background Technology

In a 1970 projection<sup>1</sup> on nuclear capacity, it was estimated that HTGRs would comprise 44% of the total nuclear capacity in the year 2000. This optimism was based on a projection of plant orders for four large reactors from General Atomic Company, with operations scheduled to begin in the period of 1981-1986. Since this time, the reactor orders were cancelled. However interest in the HTGR continues today, with the advancement of technologies in the United States, Japan, China, and South Africa.

From the very beginning, it was recognized that greater benefits of gas cooling (in particular, at that time, the ability to attain modern fossil fired steam conditions permitting, thereby, more highly efficient electricity production) would accrue if higher gas temperatures could be achieved. It was this goal, coupled with the vision that such higher gas temperatures might also lead to even broader applications of nuclear energy such as providing industrial process heat, that motivated the development of the high temperature gas cooled reactor with its characteristic reactor core of graphite moderator and ceramic fuel and its use of a gas as coolant. Figure 1 presents the evolution of HTGR technology.

Figure 1. Evolution of HTGR Technology.



<sup>1</sup> Potential Nuclear Power Growth Patterns, WASH-1098 (December 1970).

Gas cooled reactors have had a long and varied history which dates back to the very early days of nuclear energy development. Most of the early development centered on low temperature systems using a graphite moderator, metal clad metallic fuel and carbon dioxide coolant. Commercial deployment of such systems started in the mid-1950's, primarily in the United Kingdom and France, with the natural uranium fueled Magnox stations, followed by higher temperature, low enriched uranium fueled advanced gas cooled reactor stations, solely deployed in the United Kingdom, starting in the mid-1970's. Although these two pioneering programs have now concluded, experience from the over 1000 reactor-years of operation comprises a very valuable database for ongoing development and design programs on higher temperature gas cooled reactors. As of the end of 1988, a total of 52 electricity generating, carbon dioxide cooled reactor plants had been placed in operation worldwide (37 Magnox, 15 AGRs). [1,2]

To date, seven high temperature gas reactor (HTGR) plants have been built and operated. The first was the 20 MWt Dragon reactor in the United Kingdom. The HTGR came about from the combination of helium coolant and graphite moderator. Helium was used instead of air or CO<sub>2</sub> as the coolant. The graphite moderator provided enhanced neutronic and thermal efficiencies. The helium gas has a significant technical base due to the experience gained in the U.S. from the 40-MWe Peach Bottom and 330 MWe Fort St. Vrain reactors, and from Germany, which built and operated the 15-MWe AVR and the 300-MWe thorium high-temperature reactor (THTR) power plants. The German AVR operated at the 900 C level for several years. The U.S. experience was based on reactors designed by General Atomics (GA) and involved coolant temperatures below 750 C. The AVR, THTR, Peach Bottom and Fort St. Vrain all used fuel containing thorium in various forms (i.e., carbides, oxides, thorium particles) and mixtures with highly enriched uranium. [1,3,4,5]

Advancements were planned for the Ft. St. Vrain to develop three different sized commercial HTGR projects that included:

1. 2000 MWt – Summit Plant – progressed to the procurement of major components
2. 3000 MWt – Fulton Plant – design improvements completed in 1975
3. 4000 MWt – Vidal Plant – improved support structure and fuel storage design

The projects were subsequently discontinued due to technological impasses and lack of competitiveness against light water reactors. [1]

Future next generation nuclear power (NGNP) systems include modular designs that incorporate passive safety features. One high temperature reactor is being designed by an international consortium led by the South African utility, ESKOM, using a Helium cooled reactor with a direct gas turbine power cycle (i.e., Brayton cycle). This reactor builds on the high-temperature reactor (HTR) German designs using circulating graphite pebbles containing ceramic-coated oxide fuel micro particles. The design is referred to as the Pebble Bed Modular reactor (PBMR). [6]

China and Japan have built more recently smaller experimental reactors (high temperature engineering test reactor (HTTR), HTR-10) of the 10-30 MWt class, with design outlet temperatures of 900-950 C, respectively. [1,3,4,7,8,9]

The Japan Atomic Energy Research Institute (JAERI) is building on the HTTR, and is carrying out a research program on the design and development of the Gas Turbine High Temperature Reactor of 300 MWe, called the GTHTTR300. The reactor is helium cooled, graphite-moderated reactor based on pin-in-block fuel element. The GTHTTR300 has operated at 850 C core outlet temperatures. [10]

General Atomics in collaboration with Russia, is developing a gas turbine – modular helium reactor (GT-MHR) using (single phase) helium coolant, a stationary graphite moderator with high strength and stability at high temperatures, and refractor-coated particle fuel that retains fission products to high temperatures. The reactor is expected to have a core outlet temperature of 850 C. The GT-MHR development was refocused as a burner of plutonium coming from dismantled nuclear weapons. A 300 MW reactor should burn about 250 kg of plutonium per year. A first module is scheduled to be built in Russia at Seversk and to become operational in 2010-2016 timeframe. [3,7,11]

The VHTR is one of the six reactor concepts under evaluation by Gen IV. The VHTR is a thermal design that will be able to operate at temperatures at or above 1000 C, to enable production of hydrogen through thermochemical reactions for splitting water. The reactor core may be either a prismatic graphite block type core or a pebble bed. [3,7,10,12]

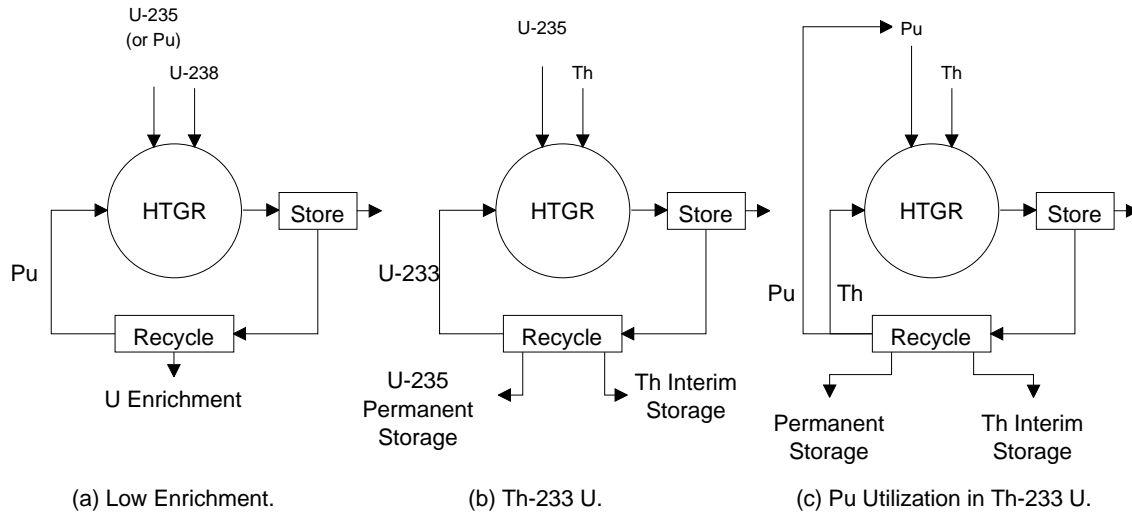
High reactor outlet temperatures are needed to drive endothermic reactions to produce hydrogen. Hydrogen is produced through thermochemical cycles that decompose water into hydrogen and recycle process chemicals. The highest priority family of cycles is the sulfur-based cycles (i.e., sulfur-iodine, hybrid sulfur, sulfur-bromine). The sulfur-iodine (S-I) cycle consists of three chemical reactions, which net the production of hydrogen. Only water and high temperature heat are input to the cycle, and only hydrogen, oxygen, and low-temperature heat are output. The minimum temperatures needed to drive this process are around 800 C, which equates to about 850 C required from the reactor to allow a 50 C drop across the heat exchanger. Reactor outlet temperatures of 950 C are expected to result in process efficiencies above 50%. [3,12]

## 2. HTGR Fuel Cycle Description

HTGRs have shown considerable adaptability to different fuel cycles. Reprocessing of the fuel would enlarge this flexibility and would even be essential for some cycles. The HTGR can use various combinations of fissile materials, U-235, U-233, and Pu and fertile materials (U-238, Th-232). HTGR fuel cycles shown in Figure 2 include:

- 1) Low enrichment, where enriched uranium fuel is burned and Pu is recycled.
- 2) Th-232, where enriched uranium and Th is burned and U-233 (and U-235) is recycled, up to the U-236 buildup limit that requires that the U-235 be retired.
- 3) Pu utilization in Th- U-233, where Pu and Th fuel is burned and Pu and U-233 is recycled. Thorium-232 is saved in interim storage for decay before refabrication.

Figure 2. Alternative HTGR fuel cycles. [14]



In the mid-1970's in Europe, the conditions governing the choice of the fuel cycle was viewed to differ from one country to another. Such conditions include: rules, codes, and construction standards; safety requirements; and definition of nuclear systems. France and Germany preferred the Th cycle compared to the U cycle. This preference was based primarily on the supply for uranium (both HEU and LEU). [13]

In 1977, it was reported that the most attractive fuel cycle for the HTGR is the Th-233 cycle. It was the only fuel cycle on which comprehensive reprocessing development work had been done. Upon completion of the planned burnup, which is usually about 90% of the U-235 present, the fuel is discharged, cooled for radioactive decay up to six months, and then shipped for reprocessing. [14,15]

In 1978, the fuel cycle that resulted in the best uranium utilization and lowest fuel-cycle costs was the HEU/Th cycle. The thorium fuel cycle was used on the Peach Bottom and the Fort St Vrain plants. The fuel consisted of fissile particles of HEU and a thorium fertile particle. In the 1970's, the conditions were favorable with low  $U_3O_8$  and fuel enrichment costs. However, the HEU created front-end proliferation concerns, so a MEU variant with 20% enriched uranium and thorium was used. This met the requirements for low fuel-cycle costs, low  $U_3O_8$  requirements, and increased proliferation resistance.

There were several recycle options possible with the MEU/Th cycle including:

- 1) Recycle only U-233,
- 2) Recycle all uranium,
- 3) Recycle all uranium and plutonium. Bred U-233 could be separated from U-235 and Pu, and denatured by mixing with U-238 so that the enrichment is in the 10-15 percent range. [1,16]

In 1997, the options cited for disposition of SNF (from HTTR, HTR-10, GT-MHR) include reprocessing, long-term repository followed by reprocessing, and direct disposal. The Japanese intention is to reprocess all spent fuel. The Russians plan to dispose fuel from the GT-MHR in a geologic repository. [8]

In 2003, the main categories of HTR fuel cycles are defined as LEU, MOX, Pu only, and Th based options. [5]

- 1) LEU cycles: uranium with an enrichment of 5-16%, recover Pu and U (still enriched to more than 1%)
- 2) MOX cycles: MOX cycles are envisaged, but little research has been conducted. Potential to have physical separation of pure Pu particles and uranium particles in reprocessing.
- 3) "Pu only" cycles: Used to consume WG-Pu and could achieve high burn-ups (500-600 GWd/MT), but would require extensive R&D program.
- 4) Th-based cycles: Uses a mixture of thorium and uranium enriched to more than 90%, would be very difficult to market due to proliferation concerns.
- 5) Th-Pu cycles: Use Pu as the only fissile material in combination with Th-232 as a fertile material. Provides better utilization of Pu vs. recycling in PWRs, and may allow replacement of the entire core at each refueling. Multiple recycling of the Pu is needed to avoid Pu buildup in spent fuels, and recycling of U-233 is beneficial.
- 6) MEU cycle: U-235 enriched to 20% (max) with thorium. Highly proliferating cycle, with incentive to recycle U-233.

Some of the issues associated with HTGR fuel cycles include:

- Fuel cycle facilities will have to be adapted to handle enrichments in excess of 5% (limit of most current facilities).
- MOX cycle has never really been studied with respect to HTGRs.
- Extensive R&D program needed to study Pu only cycle.
- Reprocessing of Th-Pu fuel enables unique advantages. [5]

### **3. Fuel (production, conversion, enrichment, fabrication)**

The fuel used in HTGRs most obviously differs from LWR fuel in that it is contained in massive quantities of graphite. Also, the fuel consists of small particles (spheres of the order of 0.5-mm diameter) of uranium oxide or carbide. The particles are coated with thin layers of pyrolytic carbon (pyrocarbon) and silicon carbide, which serve as tiny pressure vessels to contain fission products and fuel. The coated fuel particle is the basic component of the HTGR fuel element. The kernel of the coated particle consists of thorium or uranium as either a carbide or an oxide microsphere. Plutonium oxide is also an attractive fuel material. Similar fissile and fertile particles are used in both the prismatic and pebble-bed systems. [17]

The HTGR core (based on the prismatic design) consists of columns of hexagonal fuel blocks arranged in a hexagonal array and surrounded by a graphite reflector. Prototype HTGRs that used prismatic fuel were the Peach Bottom Unit 1 station and the Dragon

reactor. Both reactors use a fuel element consisting of a low-permeability graphite tube that supports a column of molded compacts containing the coated particle fuel in a graphite matrix. The graphite support tubes pass completely through the core so that fission products can be purged to a cleanup system. More recent designs for larger reactors used an unpurged fuel block. Standard fuel elements contain 72 cooling channels and 132 fuel holes to accommodate the fuel rods. [14]

HTGRs also use spherical fuel elements. The prototype reactors for this fuel were the AVR and the THTR reactor. The basic element has a diameter of 6 cm with an outer fuel-free graphite or molded carbonaceous shell and fuel particles dispersed in a matrix of graphite in the central portion. These reactors showed their high-temperature process heat capabilities by attaining outlet helium temperatures of 950 C. [14]

Two types of advanced coated fuel particles have been developed: TRISO refractory coated particle fuel and BISO. A TRISO coating contains three types of coating layers: low density pyrocarbon around the kernel to act as a buffer, and surrounding layers of silicon-carbide (SiC) and high-density isotropic pyrocarbon that retains the fission products. A BISO coating contains two types of coating layers: low-density pyrocarbon buffer and high-density isotropic pyrocarbon to retain the fission products. In large HTGRs all the HEU feed is contained in uranium-carbide ( $UC_2$ ) in the kernel of the fissile particle that has a TRISO coating. The fertile particles are BISO-coated kernels of thorium-oxide ( $ThO_2$ ). [3,4,6,12,14]

HTGRs need less heavy metal loading than LWRs at the expense of higher uranium enrichment and associated SWU demand and yellow cake consumption. Table 1 provides a comparison between the PWRs and HTGRs regarding fuel resource requirements.

Table 1. Fuel resource comparison between PWRs and HTGRs. [1,5,7,18]

|   | PWR                | HTGR             |
|---|--------------------|------------------|
| HM loading<br>(MT/GW <sub>t</sub> )                   | 26.8<br>(avg.)     | 7.5<br>(avg.)    |
| U enrichment (%)                                      | 3.0-4.2%<br>(avg.) | 14-20%<br>(avg.) |
| SWU Demand<br>10 <sup>3</sup> kg-SWU/GWY              | 135<br>(avg.)      | 221<br>(avg.)    |
| U <sub>3</sub> O <sub>8</sub> consumption<br>(MT/GWY) | 181<br>(avg.)      | 246<br>(avg.)    |

Fuel fabrication experience has been principally at General Atomics. Peach Bottom fuel consisting of mixed uranium-carbide/thorium-carbide ( $UC_2/ThC_2$ ) particles coated with a single layer of pyrolytic carbon was found to prevent hydrolysis of the  $ThC_2$  particles prior to the final fabrication. Experimentation with multiple coating layers revealed that the additional layers also provided a retention barrier for fission products. Work at Ft. St. Vrain evolved the fuel kernel coating technology. The success in fission product retention led to further development in the 1970's to separable particles of uranium and thorium to facilitate recycling of U-233. [1, 3, 4, 16, 17]

Fabrication of the fuel form consists of three major steps: kernel preparation, coating, and rod forming, after which the fuel element, comprising the graphite block and fuel rods, can be assembled. [18]

Future HTGRs will be based on U-235 fuel. The accompanying U-238 will be the source of Pu-239 (as in LWRs), which adds to the in situ fissile content. For commercial power HTGRs, the enrichment will probably be no higher than 20% in response to nonproliferation constraints. This also allows extensive generation of Pu-239, which is more beneficial in HTGRs than in LWRs. Based on studies after the Ft. St Vrain fuel composition was defined, future HTGR fuel will use a mixture of 15% carbide and 85% oxide in the fissile particles. This composition is expected to give improved fuel performance. The proved TRISO coating (over a buffer layer) will be used for future fuel. In the U.S., the prismatic block design will be used. [17]

#### **4. Fuel Management (refueling Intervals, in-core residence, burn-up)**

The HTGR is extremely flexible in that it can efficiently use several modes of operation over the lifetime of the plant:

- Non-recycle operation. Fuel charged to the reactor consists of uranium and thorium. Spent fuel removed from the core is placed in storage awaiting reprocessing and recycle.
- Initial recycle operation. An interim period for the early HTGRs when the stored U-233 is used exclusively to fuel the reactor.
- Recycle operation. The fuel removed from the core is reprocessed and the U-233 is fed back into the reactor along with enriched U-235 make-up. Another operation would be to use plutonium instead of uranium.[4]

Having chosen the overall fuel cycle, the most important independent fuel management variables are power density, fuel lifetime, and carbon-to-thorium ratio (C/Th). The combinations of fuel lifetime and power density are related to current design limits on the maximum fast fluence of about  $8.0E+21$  nvt (for neutrons that exceed 0.18 meV) and the peak fuel temperature of approximately 1350 C. The affect of these variables include:

- a) A lower power density would permit a longer fuel lifetime.
- b) For the same fuel lifetime and C/Th ratio, an increase in power density results in reduced quantity of fuel to be fabricated and reprocessed each year.



- c) An increase in C/Th (reduced thorium loading) will lead to smaller fissile inventory requirements, and smaller thorium and uranium loading will lead to reduced fabrication and reprocessing. On the other hand, the resultant smaller conversion ratio will lead to increased fuel depletion; and the lower fissile inventory will tend to give high power peaking factors and higher fuel temperatures. [4]

In a U/Th reactor, the reactor is fueled initially with enriched uranium which converts thorium to U-233. At the end of each fuel cycle the reactor operator may either sell the U-233 and start over again with enriched uranium, or recycle the U-233 back into the reactor thereby reducing the enriched uranium requirements. *In situ* fissioning of bred U-233 in the uranium-thorium cycle results in high resource utilization, particularly in the semi-homogeneous HTGR, than with any other fuel cycle. The neutronic characteristics of U-233 are superior to U-235, where U-233 yields about 10% more neutrons per absorption than U-235, and avoids the build up of U-236, a neutron poison. [4,16]

The “enrichment” of the metal in a fuel rod can be easily adjusted by appropriately choosing the relative numbers of fissile and fertile particles for a particular rod. The separate particles provide a means for axial and radial zoning of the core, thereby flattening temperature distributions while still maintaining a single uranium enrichment, as was done at Peach Bottom and Fort St. Vrain. [1]

Table 2 provides a comparison between a large PWR and a HTGR. The comparison shows that the HTGR nets greater burnup at the expense of higher enrichments. The discharged HM and Pu is significantly less than the LWR, and the HTGR has longer refueling intervals. [1,3,14]

Table 2. Comparison between a large PWR and HTGR.

|                               | Large PWR               | HTGR                   |
|-------------------------------|-------------------------|------------------------|
| Burn-up                       | 33- 50 GWd/MT<br>(avg.) | 83-112 GWd/MT<br>(MEU) |
| Discharged HM<br>(MT/GWY)     | 21.4<br>(avg.)          | 5.4<br>(avg.)          |
| Discharged Pu<br>(kg/GWT)     | 235<br>(avg.)           | 109<br>(avg.)          |
| Discharged Pu-239<br>(kg/GWY) | 171<br>(avg.)           | 43<br>(avg.)           |
| Refueling Interval            | 12-18 months            | 12-24 months           |

JAERI is developing a two batch axially shuffled “sandwich” refueling scheme for the GTHTR300. This scheme is combined with some fully inserted power rods and burnable poisons ( $B_4C + C$ ) of large diameter and low boron content to keep the total reactivity variation minimum throughout the burnup period. Spent fuel is unloaded from alternate axial layers and the remaining fuel blocks are shifted downward while the new fuel is reloaded in the vacated layers. [10]

General Atomics is proposing a novel concept called Deep Burn transmutation that is based on the use of thermalized neutrons and high burnup fuel forms in MHRs. In the deep burn concept, the thermally fissile component of the nuclear waste is used as the fissionable “driver” fuel to generate the neutrons necessary to achieve the conversion of the thermally nonfissile component (i.e., transmutation fuel) into fissile isotopes that can be destroyed. The transmutation fuel also functions as a burnable poison and provides reactivity feedback control. In this concept, the destruction of the TRU reactor waste is carried through one burnup cycle, achieving complete destruction of weapons-usable materials, and roughly 95% destruction of all TRU waste, with only one intermediate reprocessing step that does not involve significant amounts of weapons-usable material.

More than 75% overall destruction of the TRU can be achieved in special purpose reactors using MHR technology, operating as critical systems. To extend the destruction of the TRU beyond 75%, the irradiated transmutation fuel is extracted from the MHR critical system and is then irradiated without further reprocessing in a second-stage subcritical system, thereby avoiding the problems associated with the handling of volatile higher actinides (Am and Cm).

These reactors have annular graphite-moderated cores, and are designed to be passively safe at power levels up to 600 MW, such that there is no fuel failure or fission product release under any loss of coolant flow or pressure accident. An essential feature of the deep burn transmutation concept is the use of TRISO fuel particles that allow deep burnup without multiple reprocessing. [9].

## 5. Reprocessing

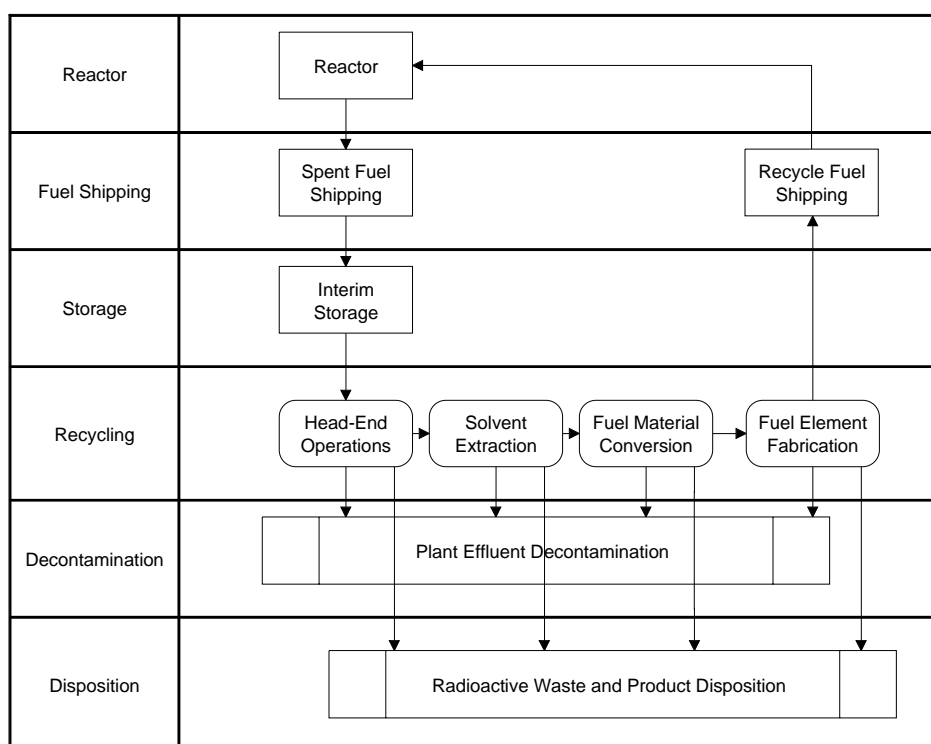
In spite of numerous research works of R&D in the past, there is not yet an available process for the treatment of HTGR spent fuels today. While such a process is entirely feasible, there is no existing, readily available process technology such as for LWR fuel. However, there have been significant cold and some limited-scale hot engineering tests of these processes during the 1960-1970's. [3,5,14]

The difficulties of the reprocessing of the HTR spent fuel are bound to the nature of its constituents. The graphite and the silicon carbide are chemically inert compounds and few chemical reagents are capable of dissolving them quantitatively and effectively. These difficulties are also bound to its structure. The volumic fraction of the kernel containing Heavy Metals (HMs) to recover (fissile and fertile isotopes) is very small. It is lower than 1% in the case of the pebbles of the PBMR concept. The small dimension of

these kernels does not allow us to envisage a simple step of shearing to reach the HMs like for the reprocessing of the LWR spent fuel. [5]

There are several strategies considered for recycling HTGR fuel. The strategies range from no recycle, to mixed recycle, once through recycle, to full recycle. Many of the cycles involve in some way the recycle of bred U-233. In reprocessing, the BISO and TRISO can quite successfully be separated for further disposition. U-236 poisoning is avoided by limiting the recycle of uranium feed remaining in the discharged fuel. The fuel recycle process generates waste and product streams for disposition as shown in Figure 3. [4,20,21,23]

Figure 3. Major fuel reprocessing operations and waste effluents.



In U-233 and thorium reprocessing, the SNF is received from the reactor and is temporarily stored. The total decay time prior to reprocessing will permit essentially complete decay of Pa-233 to U-233, in about 120 days. The fuel reprocessing sequence starts with the head-end process, where the fuel in the HTGR fuel element is separated from the graphite body. The SNF elements are first prepared for burning by crushing (primary burner feed preparation) or by milling. Primary burning eliminates most of the moderator and the outer coatings of particles. Particles are then classified to separate the U-235, thorium, U-233, and fission products. Particles having silicon carbide coatings and intended for recycle are then crushed and burned in a secondary burner. The metal oxide ash is dissolved to create a solution of uranium, thorium, and fission products; the silicon carbide and coated U-235 remain as a residue. The U-235 is separated mechanically and the uranium and thorium are individually recovered from the fission

products. The recovered U-233 and thorium are stored for reuse as fuel. Large-scale operations are involved in the reprocessing flow sheet to handle off-gases and the liquid and solid waste. A detailed description of the reprocessing steps may be found in the following references. [4,8,15,18,20,21,22,23]

During the mid-1960's, an extensive national program plan for reprocessing and refabrication of U/Th fuels was developed by ORNL, and GA. This effort was a part of the "National HTGR Fuel Recycle Development Program Plan,"<sup>2</sup> The impetus for the program was to conserve uranium resources while supporting nuclear power growth. A cold engineering-scale pilot plant was installed at GA (1970), and several years of development of HTGR reprocessing unit operations (fuel element crush, burn, dissolution, solvent extraction, and off-gas retention) were conducted. [17,18]

As part of the joint U.S./Federal Republic of Germany (FRG) program, a small-scale hot pilot plant was constructed in Germany. The Jupiter Experimental Reprocessing Facility was developed to experimentally reprocess AVR fuel elements. The Jupiter project was started in 1971 and was designed as a model to gain experience and data that could be used later in the design of a bigger plant. The plant would operate at 2 kg HM/day and includes graphite combustion, dissolution of the fuel particles in Thorex reagent, and solvent extraction. [24]

Head-end treatment of HTGR-type fuels was done at the Idaho Chemical Processing Plant (ICPP) for nuclear rocket fuels. After successfully operating a pilot plant burner in 1970, design and construction of a larger prototype burner was started in 1972. This facility went through cold testing. Hot testing was scheduled for 1982. [16]

The French recognized that reprocessing and refabrication, to be economically viable, requires a large-scale capacity. In 1974, France considered adapting existing facilities (i.e., UP1 at Marcoule) for the reprocessing of HTR fuels. The advantages of such an operation were that existing reprocessing lines could be adapted for thorium-based fuel reprocessing at little extra cost; the installation of a head-end would benefit from the existing logistical support; and the Marcoule site should pose no problems for the extension of the existing plant to include the head-end process and the fuel element refabrication plant. [13]

The National HTGR Program resulted in the following technologies:

- Head end- graphite crushing and burning, particle crushing secondary burning, and ash dissolution have been carried out in cold engineering-scale studies. Also some confirmatory work on burning and ash dissolution has been done on irradiated specimens in small-scale hot cell work. However, problems remain to be solved in developing a satisfactory graphite burner and solids handling equipment.
- Solvent extraction- no major developments on Purex or Thorex Process required because operations are not significantly different than have previously been conducted in plant operations. The TRUEX process had only been developed on

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<sup>2</sup> National HTGR Fuel Recycle Development Program Plan, ORNL-4702. Rev. 2, circa 1976.

a laboratory scale. Pilot scale work in hot cells will be required to develop and demonstrate the process.

- Off-gas treatment-methods exist for the removal of the various radioactive gases but development will be required for an integrated treatment system.
- Plant design- considerable effort and lead time will be required to develop a plant design and associated database that meet present day environmental, health, and safety requirements imposed by state and federal agencies. [17]

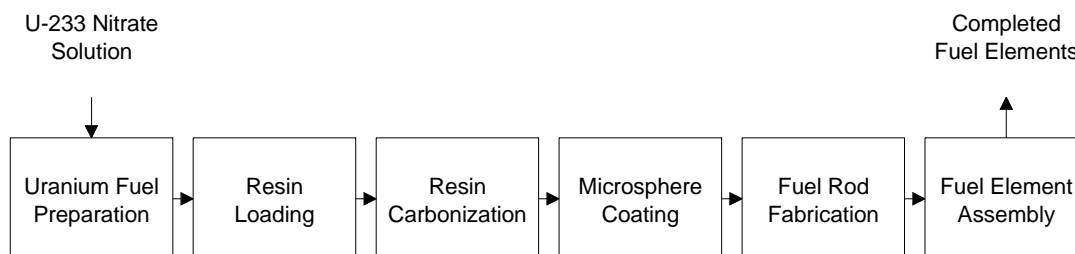
## 6. Refabrication

The prime distinction between “fabrication” of fresh fuel and “refabrication” of recovered fuel is that the latter is done with radioactive (or hot) materials and must, therefore, be performed remotely in a hot cell. The two major purposes of reactor fuel reprocessing are to recover and purify fuel and to convert the wastes to a form suitable for disposal.

U-235 must be extracted from the SNF elements, including partitioned from the purer (less U-236) bred U-233, and then refabricated into special dedicated fuel elements for further irradiation. This refabrication must be accomplished in the remotely operated, and expensive, refabrication portion of the recycle facility because of the U-232 which may cross over from the bred fuel to contaminate the residual U-235 recycle material. [20]

In refabrication, U-233 is introduced from reprocessing storage, it is decontaminated by ion exchange if necessary, and the solution is adjusted chemically for loading onto ion exchange resin. The loaded particles are carbonized, converted to the proper stoichiometry, and then coated with various layers of pyrolytic carbon and silicon carbide. Following particle coating, the fissile particles are mixed with coated fertile particles prepared in contact facilities and fed to the fuel-rod fabrication step, where the particles are bonded together with a carbonaceous matrix. The fuel rods are placed into a premachined graphite fuel block, and the complete fuel assembly is then cured in place at high temperature. Substantial operations in the refabrication flowsheet are involved with scrap treatment and waste treatment for off-gases, liquids, and solids. A simplified flow sheet is provided in Figure 4. [4, 15,18,20,21,22,23]

Figure 4. Process flow for spent fuel refabrication.



For a detailed description of the refabrication process including nitrate solution, sol-gel microspheres preparation, drying and calcine, microsphere coating, blending, fuel rod preparation, carbonize, fuel element preparation, and waste disposition see references by Brooks. [18,21]

## **7. Fuel handling, interim storage, shipping casks, and transportation**

Fuel handling encompasses shipping and storage of spent and refabricated fuel elements. A surge capacity must be available at the recycle plant since the plant must operate on a sustained basis. HTGR fuels were planned to be stored for up to six months prior to shipment to a recycle plant. Due to the long fuel cooling times and low power density of HTGR cores, fuels pose no severe problems of fission product decay heat removal during shipment. [14]

The spent fuel must be stored in a shielded facility that can handle the low-level (but not inconsequential) heat load, has the capability of retaining the identity of individual fuel elements, and has the capacity to store certain types of elements until a campaign can be run. The identification of elements is important in terms of fuel type and ownership due to several utilities shipping fuel to a reprocessing plant. [23]

In 1972, the HTGR SNF shipping system was developed to provide the method and equipment required for removing and transporting SNF elements from a nuclear plant to a storage and/or reprocessing plant. The system must be designed such that all federal regulations and DOT, in addition to applicable state laws are satisfied. The two approaches to shipping are by rail and by road.

- 1) The rail-shipping package consists of a cask and twelve containers. Each container holds six spent or five refabricated fuel elements. The cask is fabricated in three layers: a stainless steel inner layer, a middle layer of depleted uranium, and an outer shell of steel. The cask weight is 160 tons and it is transported in the near-horizontal position on a special railcar weighing 60 tons. The cask is finned for improving the removal of decay heat generated by the fission products. The cask is not removed from the car for loading/unloading operations but is jacked into an upright position when the lid can be removed and the fuel transferred.
- 2) Truck shipping differs from rail shipping in the size of the cask. The truck-mounted cask will accept one container that is identical to the rail container. [18,21]

Fuel storage of Fort St. Vrain HTGR and Peach Bottom core segments was performed at the Irradiated Fuel Storage Facility (IFSF). The fuel was stored in steel canisters in a storage vault, and cooling was provided by forced convection of filtered (inlet and exhaust) atmospheric air. The storage vault capacity was about 2500 fuel elements. A commercial scale facility would need a capacity of about 10,000 to 20,000 fuel elements, cover an area of about 1-2 acres and provide a heat dump capable of absorbing 4000 kW. [14,16]

The fuel storage system concept for the GT-MHR fuel includes the use of multi-purpose canisters (MPC), which could be used for storage, transportation, and permanent disposal of the SNF. The GT-MHR MPC would contain 42 fuel hexagonal graphite elements of 0.8 m length, and 0.36 m across the flats, arranged as seven columns with six fuel elements per column. Each fuel element would contain about 20 million fuel coated particles. [8]

The size of the reactor and the distance between the reactor and the recycle facility are the two most important considerations in selecting the mode of transportation for the SNF. Cost studies indicate that the most economical method of shipping spent HTGR fuel for an 1160 MWe reactor is in a 150-160 ton railroad cask. The handling and shipping of spent and recycle HTGR fuel elements is quite similar. All handling operations are accomplished by remotely operated equipment. The same fuel containers and fuel shipping cask are used to transport both types of fuel elements. [4]

## **8. SNF waste forms**

There are several types of wastes that are generated in the HTGR fuel cycle. The wastes that result from reprocessing of SNF may take several forms: aqueous, solids, graphite moderator blocks, and spent fuel. Many of the wastes from reprocessing could be converted into glass to meet acceptance criteria of the repository. [17]

Aqueous waste: Aqueous wastes from the various fabrication process steps are collected in hold tanks, monitored, concentrated by evaporation, packaged in drums, and shipped to an off-site disposal area for burial. [21]

Solid waste: Solid wastes consist primarily of the graphite sleeves used as liners in the carbon coating furnaces and off-specification coated particles. The sleeves contain small amounts of U-233. The graphite liners from the coaters are moved in a transfer container to a crushing machine, crushed, sampled, counted, and then burned. The uranium containing ash is leached with nitric acid and shipped back to the reprocessing plant for purification. [21]

Graphite blocks: If whole block spent fuel is unacceptable for repository emplacement; the first processing option is to separate the spent fuel into (1) the spent fuel matrix material and (2) the carbon from the graphite block. The fuel rods could be packaged for disposal at the repository or the separated fuel could be burned and chemically processed into final products consisting of fissile-fertile byproduct and suitable waste form for the fission products and actinides. Acceptable waste forms could include overpacks, coating, or encapsulation technologies. [3,8,25]

Waste forms might be improved by:

- 1) Blocking coolant channels with graphite plugs. Air and water reactions with graphite are surface phenomena. Most of the surface area of a fuel block is in the coolant

channels. If the coolant channels are plugged, the surface area (and area for chemical reactions) is reduced by 77%.

- 2) Coating the graphite block with a protective surface coating. There has been limited research in the reactor community to improve chemical resistance of HTGR fuel blocks under severe accident conditions –typically air at temperature of ~ 1600 C. Coating options include ceramics such as silicon-carbide (SiC). Such coatings may also reduce long-term oxidation rates. [17]

Spent Fuel assemblies: Scoping calculations suggest HTGR spent fuel is a superior waste form to LWR spent fuel. This should simplify licensing and may reduce cost per unit volume for the waste package. [17]

The PBMR fuel spheres (TRISO) provide a multi-barrier for containment, given that the stable silicon carbide layer around the uranium will last for more than a million years and the coal-like graphite itself does not degrade underground or within running water. [1,3,6]

## 9. Waste treatment

HTGRs produce less heavy metal radioactive waste per unit of energy produced because of the plants high thermal efficiency and high fuel burnup. Additionally, less total plutonium (proliferation concern) per unit of energy produced. [3]

Table 3 shows the advantage that HTGRs have over LWR plants relative to waste heat. The thermal discharge from a HTGR is less than a LWR plant because of greater thermal efficiency. The HTGR requires less than 60% (compared to LWRs) of the water coolant per unit of electricity produced. Because of the lower heat output, the waste heat can be rejected directly to the atmosphere using air-cooled heat rejection systems, thus avoiding use of water coolant resources. [3]

Table 3. Waste heat requirements for LWRs and HTGRs.

|  | Large PWR | HTGR |
|--|-----------|------|
| Heat rejection (GWt/GWe)                     | 1.8       | 1.1  |
| Cooling water required ( $10^4$ acre-ft/GWy) | 2.4       | 1.4  |



There are three radioactive waste treatment systems for the HTGRs: the gaseous, liquid and solid radwaste treatment systems. All potentially radioactive gases, liquids and solids are collected and processed according to their physical and chemical properties and radioactive concentrations. The radwaste treatment systems collect and monitor all potentially radioactive wastes from the plant. [9]

Gaseous Treatment. Gaseous effluents from the reprocessing plant will be treated to remove radioactive and non-radioactive pollutants. A major component of gaseous radioactive waste is the carbon dioxide from the burners. These gaseous wastes will contain Kr-85, I-129, and H-3, non-radioactive gases, and suspended particles. A combination of filtration and the KALC process was developed by ORNL to clean up this gaseous waste. [18]

Storage of Kr-85 (half life of 10.7 years) and H-3 (half life of 12.4 years) for about 100 years would permit adequate decay for subsequent release. Storage of I-129 (half life of  $1.7 \times 10^7$ ) would be required for essentially perpetuity. Other wastes include the fission products and long-lived actinides. The former, which are beta and gamma emitters, will be concentrated, dried and fired, and converted to a storable form (e.g., by encapsulation in a glass matrix). The latter, whose primary hazard is that they include alpha emitters, are given the same treatment as the fission products, but consideration is being given to separating out the actinides and giving them special treatment. [23]

The head-end processes may release C-14 to the atmosphere that may require additional treatment. For example, the  $\text{CO}_2$  released by burning could be converted to stable  $\text{CaCO}_3$  by reaction with lime, and then stored as a low-level waste. [23]

Liquid Treatment. Liquid effluents will be concentrated by evaporation and added to the high-level liquid waste stream for calcinations to a solid. Solids from the calciner will eventually be canned and sent to a federal repository. [18]

Solids Treatment: Some HTGR recycle wastes are not significantly different from LWR fuel recycle wastes, and thus can be processed and isolated similarly. HTGR recycle wastes that are unique and not encountered with LWR wastes include silicon carbide hulls, retired U-235 fissile particles, fluoride in the solvent extraction waste, and C-14 that is greatly diluted by large quantities of  $\text{CO}_2$ . [14,23]

Graphite blocks may not be incinerated due to the C-14 problem, but could be treated to improve the properties for disposal (or possibly recycling). Potential treatments include thermal/chemical treatment to remove absorbed radionuclides (solvents, gases), or by coating (e.g., metal, ceramics). [25]

## 10. D&D issues

Experiences in Decontamination and Decommissioning (D&D) of HTGR have created a basis for how to design from the beginning to make D&D simple.

The 15 MWe AVR experimental reactor was shut down in 1988, and decommissioning began in 1994. Since a pebble bed reactor is never defuelled during reactor operation, defueling is the major concern in Safestore<sup>3</sup> decommissioning, and the whole task was separated in a first phase with defueling and dismantling outside of the reactor building and a second phase with dismantling and preparations for the later dormancy period inside the reactor building. [25]

The Fort St. Vrain nuclear generating station had been in commercial operation for over a decade prior to its permanent shutdown in 1989. The HTGR state was subsequently defuelled and decommissioned with the plant site released for unrestricted use in August 1997. The decommissioning method selected was “early plant dismantlement” rather than the 60 year Safestore option. The removed fuel was ultimately stored on-site at the Independent Spent Fuel Storage Installation. [8]

Decommissioning plans for China’s HTR-10 reactor were developed in its design, as required by regulation. The HTR-10 reactor adopts the modular high temperature reactor design. It uses steel pressure vessels and the RPV and the SGPV are arranged in a “side-by-side” manner. They are separately housed in two concrete cavities. The spherical fuel elements can be easily removed from the reactor core into containers without opening the RPV. These factors, among other design features, make the decommissioning of the HTR-10 relatively easier. [8]

## **11. Disposal (inc. waste packages)**

Waste acceptance criteria for the presently planned underground waste disposal facilities have been conceived primarily with the large volume waste forms in mind, although it has been recognized that there are a number of less common forms of spent fuel that must be accommodated. While the criteria were made quite general to cover as many unanticipated situations as possible, the special case of spent HTGR fuel was not specifically considered.<sup>4</sup> Options for disposal of HTGR fuel in a repository include whole-block disposal, disposal with removal of graphite, and reprocessing of spent fuel to separate the fuel and fission products. [10,17]

Given the quantities of the currently existing HTGR SNF, the whole block disposal of the fuel is not expected to have a major impact on repository capacity unless a large-scale deployment of HTGR technology is undertaken. Estimates of the number of FSV canisters range from 200-700 depending on canister size. Comparatively, there may be 15,000 canisters of vitrified HLW and 45,000 canisters of LWR spent fuel. [17]

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<sup>3</sup> ‘Safestore’ is a process, which the decommissioning activities are carried out in a number of steps separated by quiescent periods of care and maintenance.

<sup>4</sup> “Design Criteria for the Waste Package,” 10 CFR 60.135, Criteria for the Waste Package and Its Components, (1991), paragraph (3).

HTGR fuel, if disposed as a whole fuel element, has the disadvantage of requiring considerably more volume for storage of a unit weight of fuel and fission product isotopes. A typical waste canister for the Yucca Mountain repository is sized to contain a mix of pressurized water reactor (PWR) and boiling water reactor (BWR) assemblies equivalent to a spent fuel quantity of about 1 MTIHM. By comparison, an equivalent waste canister would contain a vertical stack of four fuel blocks (Fort St. Vrain type), or approximately 40 kg of heavy metal. Thus, improvements in the fuel design and performance to enhance the feasibility of separation of graphite from the spent fuel should be considered. [17]

On the basis of C-14 concentration, the graphite block has the potential, dependent upon fission product contamination, of qualifying as Class C LLW (concentration limit of 8 Ci/m<sup>3</sup>). If graphite waste from an HTGR fuel cycle is classified as LLW, then near-surface disposal may be an option. If the graphite block is not separated from the spent fuel, the spent fuel elements must be disposed at the proposed repository. If direct disposal is not allowed, the option exists to “overpack, coat, or encapsulate” whole HTGR spent fuel blocks to improve disposal performance of the waste before packaging. [17]

HTGRs have reduced risk of repository SNF radionuclide migration to the biosphere. The PBMR fuel spheres are already an ideal container, given that the stable silicon carbide layer around the uranium will last for more than a million years and the coal-like graphite itself does not degrade underground or within running water. [3,6]

If only small quantities of HTGR spent fuel are to be disposed, then standard waste canisters would be used. If there were large-scale deployment of HTGR technology, part of any repository would be optimized for disposal of HTGR spent fuel. Two reasons have been cited: 1) Repository cost is primarily controlled by decay heat load. An LWR has a power plant efficiency of 32 to 35% vs. 38 to 40% for a HTGR. The 20% greater power plant efficiency of the HTGR implies ~20% less decay heat in SNF per unit of electricity generated. Twenty less decay heat per unit of electricity generated implies 20% fewer tunnels required in the repository per unit of electricity generated to spread out the heat level underground. 2) The geometry of the HTGR SNF assembly allows a more optimized waste package/repository design than for LWR spent fuel with significant cost savings per unit volume. [17].

Intermediate options for reoptimizing waste packages include: taller canisters (60 kg/canister), larger diameter canisters (420 kg). Given the lower thermal density of HTGR fuel, it should be possible to decrease the distance between boreholes containing HTGR fuel, thus increasing repository capacity. This option needs further study in order to assess its feasibility and advantages.

## 12. Future Research and Conclusions

The HTGR fuel cycle is characterized by high burnup, low power peaking factor, extended refueling interval, difficulty in reprocessing HTGR fuel kernels, U-233 fabrication, graphite block waste form, Carbon-14 issues, and lower decay heat. These challenges extend beyond LWRs, but draw on many commonalities in fuel processing, fabrication, waste treatment, waste forms, transportation, and disposal. Disposition strategies are needed for legacy HTGR wastes and for future Generation IV gas reactors. A summary of the research opportunities for each of the fuel cycle elements is provided in Table 4.

Table 4. Research opportunities by fuel cycle element.

| <b>Fuel Cycle Element</b>    | <b>Research Opportunities</b>  |
|------------------------------|--|
| 1. Fuel                      | TRISO fuel design, higher enrichments (than LWRs) and high burn-up fuels, fuels for prismatic block and pebble bed designs, U/Th fuels, Plutonium, and transmuter fuels. |
| 2. Fuel management           | Refueling schemes, deep burn transmutation   |
| 3. Reprocessing              | Engineering scale reprocessing of HTGR fuels, graphite burning & solids handling, separation of graphite from SNF.   |
| 4. Refabrication             | TRISO and BISO fuel refabrication with U-233. Refabrication with Pu, and transmuter fuels.   |
| 5. Fuel handling             | Packing of graphite fuel   |
| 6. SNF waste forms           | Graphite block waste forms, waste package design for optimal disposal.   |
| 7. Waste treatment           | Integrated waste treatment systems, unique HTGR wastes, C-14 issues.   |
| 8. D&D                       | D&D as part of the original reactor design concepts, D&D of high temperature materials, thermo-chemical process D&D.   |
| 9. Disposal & waste packages | Graphite block reclassification as LLW, implications on repository capacity from less decay heat and less HM.  |
| 10. Total fuel cycle         | Modular design and economics of production. Use of the MOX cycle, Pu-only, and Th & Pu, and U/Pu/Minor Actinide cycles.  |

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